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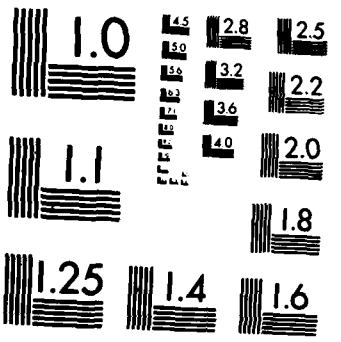

OPERATING CONDITIONS FOR THE HYDROGEN/DEUTERIUM TRIPLET LASERS

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Abstract

Research on energy storage and transfer in molecular hydrogen and its isotopes lead to the discovery of mid-infrared lasers in H_2^1 and H_2^2. Specifically, the laser lines in H_2^1 are at 3.84, 3.77 and 3.71 μm; in H_2^2 they are at 4.71, 4.60, and 4.52 μm. These have been reported by us recently(1,2,3). We conclude that these lines in both isotopes belong to the $a^3\Sigma_g^+ - c^3\Pi_u$ transitions. For H_2^2 there is still some doubt since several of the spectroscopic coefficients for the H_2^2, $c^3\Pi_u$ state have not been determined. In this report we describe the conditions under which the lasers occur and show how this leads to a plausible explanation for their existence. We derive several plausible sets of value for the spectroscopic coefficients for the H_2^2 $c^3\Pi_u$ state.

Introduction

Research on energy storage and transfer in molecular hydrogen and deuterium led to the discovery of mid-infrared lasers in these gases. Specifically, laser lines at 3.84 μm, 3.77 μm, and 3.71 μm in hydrogen and at 4.71 μm, 4.60 μm and 4.52 μm in deuterium were identified and reported(1,2,3). Our conclusions are that these lines are part of the $a^3\Sigma_g^+ - c^3\Pi_u$ series for both isotopes of hydrogen. For normal hydrogen this seems to be certain. For deuterium there is still some doubt since several of the spectroscopic coefficients for the deuterium $c^3\Pi_u$ state have not been determined(4). In this report we describe the conditions under which the lasers occur and show how this leads to a plausible explanation for their existence: A tentative set of spectroscopic coefficients for the deuterium $c^3\Pi_u$ state are given.

Laser Apparatus:

The lasers were discovered during a search for weak or induced dipole lasers in molecular hydrogen, deuterium, and deuterium hydride(5). A detailed description of the laser apparatus has been previously reported(1). The laser basically was a glass tube 6.3 m long and 2.5 cm diameter with adjustable internal mirrors which were highly reflective in the laser bands and nearly transparent for visible light. The mirrors were dielectric coated and "tuned" to the desired infrared bands with output mirrors having 80% to 90% reflectivity and the back mirrors having greater than 99% reflectivity. Cavity alignment was verified by a helium-neon laser.

The lasers were excited by a four-stage Marx bank at erected voltages of 225 KV to 320 KV. Current limiting resistors were placed in series with the discharge to prevent discharge collapse and oscillation. Gas pressures were varied from about ½ torr to 10 torr, and two gas temperatures were used: ambient (∿ 300°K) and cryogenic (∿ 100°K). Variations in voltage, series resistance, gas pressure, temperature and gas composition all influenced the laser's output characteristics. Figure 1 is a sketch of the laser apparatus. Figure 2 is a photograph of the laser in the laboratory. Figure 3 is a picture of the high voltage end of the laser showing the current limiting resistor, peaking gap, corona ring and cryogenic jacket.

Data/Data Reduction:

The following parameters were measured directly or inferred from direct measurements: 1) initial voltage, 2) voltage vs. time, 3) current vs. time, 4) gas composition, initial temperature and pressure, 5) total laser output vs. time, and 6) spectrally resolved laser output vs. time. Figure 4 shows the voltage, current and detector signal in a typical experiment. Figure 5 shows the spectrally resolved components of the laser pulse. In many cases, as shown here, an identifiable time delay (∿ .2 μ sec.) occurs between the onset of the strong line at 3.77 μm and the other two lines. Additionally, in order to facilitate data reduction, a data field of voltage, current and initial gas pressure was obtained

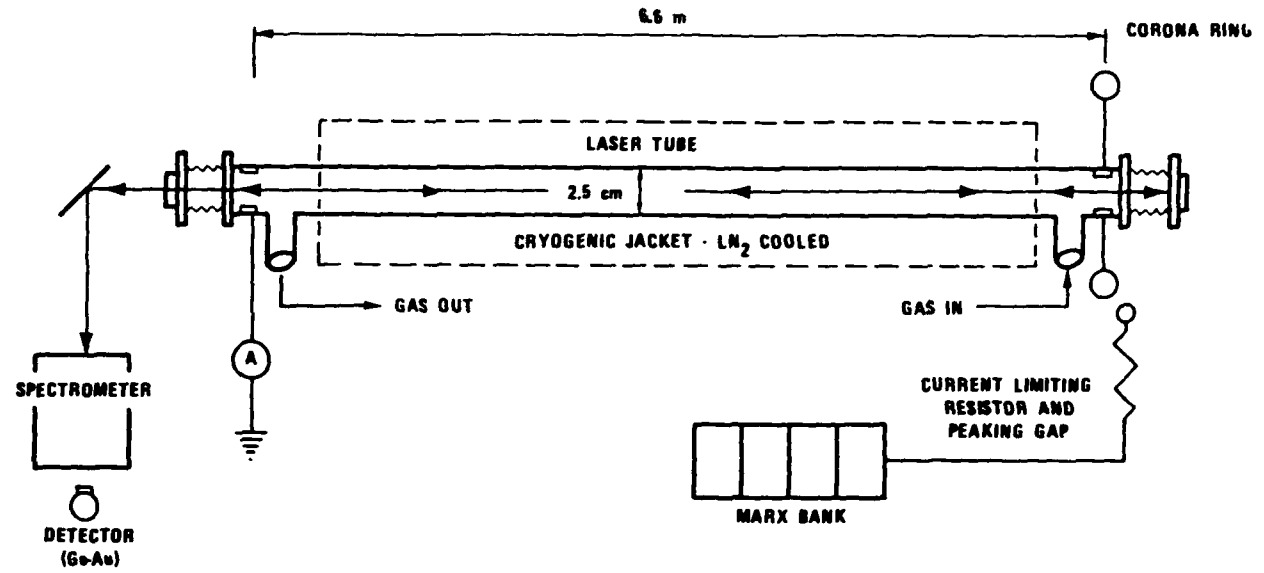

Figure 1. Apparatus for the hydrogen and deuterium triplet lasers.

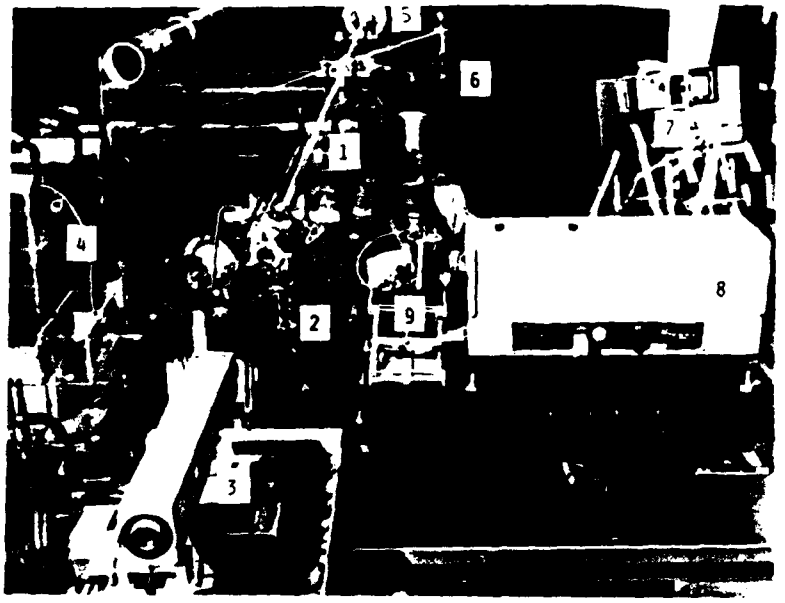

Figure 2. Hydrogen triplet laser in the laboratory.
1) Laser tube (6.6 meter length x 2.5 cm diameter) on a wooden optical bench.
2) Output window on sealed adjustable mount.
3) HeNe alignment laser.
4) Four-stage Marx bank.
5) Corona ring for high voltage end (detail in Figure 3).
6) 10:1 voltage divider.
7) Storage oscilloscope.
8) 3/4 meter grating spectrometer, and
9) Cryogenic gold doped germanium infrared detector.

for hydrogen at ambient temperature. At high currents (i.e., high voltage and/or low series resistance) the discharge would become an arc which placed an upper limit on current. At very low currents the excitation was insufficient to produce laser action.

The instantaneous state of the electric discharge was determined from the measured parameters and the electron drift velocities published by Englehardt and Phelps(6) or more recently calculated drift velocities from Taylor(7) based on the Boltzman transport code developed by Carleton and Megill(8). Computation of variables proceeded as follows:

a) Laser action occurred only during the glow discharge. This type of discharge was estimated to fill approximately 2 cm out of the 2.5 cm bore of the tube. Discharge cross section was taken to be ~ 3 cm^2.

b) The voltage measured included both the potential drop in the gas and the electrode potential drop. In this work the electrode potential drops were assumed to be small so, to a first approximation, the entired measured potential was assumed to be across the gas column.

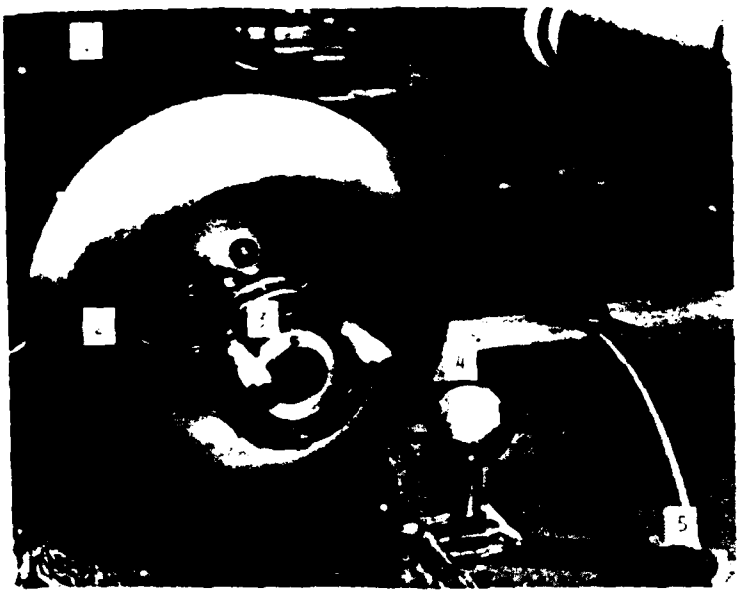

Figure 3. High voltage end of the laser in the laboratory.
1) Cryogenic jacket.
2) High voltage corona ring.
3) Concave back mirror on adjustable sealed mount.
4) High voltage discharge peaking gap, and
5) Current limiting resistor.

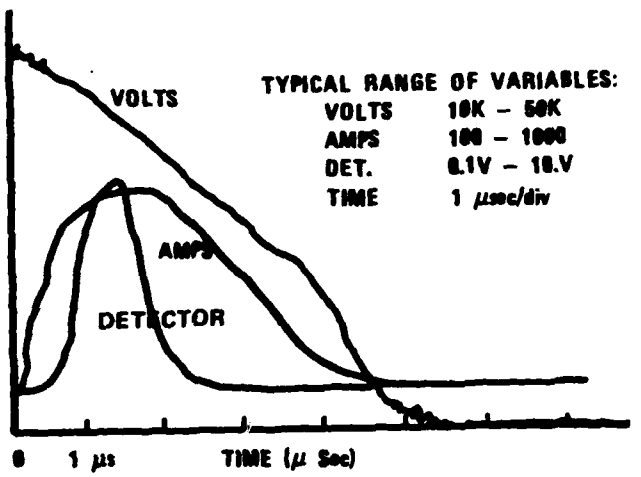

Figure 4. Typical voltage, current and detector outputs (from oscilloscope traces)

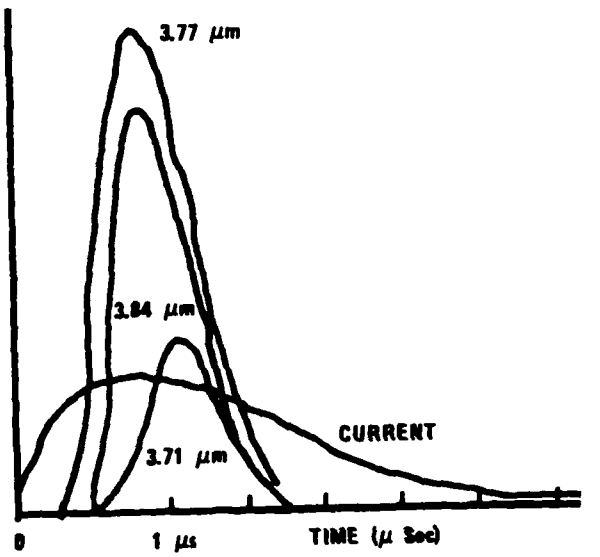

Figure 5. Spectrally and temporally resolved lines of the hydrogen triplet laser.

c) With the voltage, discharge path, and gas density known, values for the electric field strength (volts/cm) and for the electric field per particle, E/N volt cm^2 could be determined. Then using transport data from Englehardt(6) or Taylor(7) the electron drift velocity could be obtained from their published calculation for a given E/N in hydrogen gas. From the same sources the characteristics electron energy (electron volts) was also obtained.

d) From the measured current, known discharge cross section, and the electrons drift velocity, the concentration of conduction electrons was calculated.

e) The calculated electron concentration and the characteristic electron energy can be used to calculate an electron energy distribution. The simplest thing at this point is to assume a Maxwell-Boltzman energy/velocity distribution for the electron.

f) Finally, the cross section data from Böse(9) for the electron collision excitations of hydrogen from ground to the first triplet state can be used with the available free electron concentrations in the desired energy ranges to calculate the rate of production of $a^3\Sigma g^+$ and $c^3\Pi u$ states.

TABLE I

HYDROGEN TRIPLET LASER OPERATING CHARACTERISTICS

Laser Cross Section: 3 cm^2

Length: 6.6 m

Volume: 2.1 ℓ

Series Limiting Resistor: 100 Ω

Gas Density: $6.5 \times 10^{16}/cm^3$ (hydrogen)

Initial Pressure: 2 torr (T, ambient)

TIME (μ sec)	V# (volts)	I (AMPS)	E v/cm	E/N $v\text{-}cm^2$	ω* cm/sec	εk+ e volts	N cm^{-3}	P $watts/cm^3$	∑P Δt $joules/cm^3$
0	8.E4	0.	121.	1.88E-15	1.8E7	3.8	0	0	0.0
0.5	7.6E4	360.	115.	1.78E-15	1.7E7	3.6	4.22E13	1.3E4	6.5E-3
1.0	7.3E4	520.	111.	1.72E-15	1.6E7	3.3	6.5E13	1.8E4	1.6E-2
1.5	7.0E4	580.	106.	1.64E-15	1.5E7	3.2	7.7E13	2.0E4	2.6E-2
2.0	6.5E4	520.	99.	1.53E-15	1.4E7	3.1	7.4E13	1.6E4	3.4E-2
2.5	6.0E4	420.	91.	1.41E-15	1.3E7	3.0	6.4E13	1.2E4	4.0E-2
3.0	5.6E4	300.	85.	1.31E-15	1.2E7	2.8	5.0E13	0.8E4	4.4E-2

LASER: Start 0.9 μ sec
Peak 1.5 μ sec (detector output, 6 volts), (9.9 Kj/ℓ-atmos input)
End 2.4 μ sec

Data Ref: Case B, 6 August 1982

\# E4 = 10^4, etc.
* Electron drift velocity - current carriers
\+ Electron characteristic energy (electron "temperature")

The bottoms of the potential wells for these states are 95936.1 cm^{-1} for the "a" state and 95838.5 cm^{-1} for the "c" state. The "a" and "c" state potential curves are almost identical; for all practical purposes they have the same Franck-Condon characteristics. We, therefore, assume that they are nearly equally populated. Table I shows representative samples of the current and voltage data obtained and reduced according to the above technique.

Gain and Laser Output:

The gain of the hydrogen and deuterium triplet lasers derives from the ortho-para population distribution in the rotational states. In hydrogen the ortho-para population ratio is 3 to 1 in rotational equilibrium with the highest population being in the odd numbered rotational states(10). In deuterium the ortho-para ratio is 2 to 1 in rotational equilibrium with the highest population being in the even numbered rotational states. In deuterium hydride there is no ortho-para asymmetry and therefore, no potential for laser gain.

We have calculated the populations for various vibration-rotation combinations of the $a^3\Sigma g^+$ and the $c^3\Pi u$ electronic states assuming thermal equilibrium within the rotational state and equal total state

populations. Temperature of 100°K and 300°K were assumed. At 300°K in hydrogen the favored rotational transition is from J = 1 to J = 2, that is, this transition has the highest population ratio. This matches conveniently the hydrogen line assignments based on measured and calculated wavelengths. For deuterium the case is not so clear. Our simple rotational equilibrium modelling shows that the J = 2 to J = 3 transitions are somewhat more favored than the J = 0 to J = 1 transition at 100°K and higher. In spite of this, it appears that the J = 0 to J = 1 transitions are the ones that are running in the deuterium laser.

Calculated and Measured Wavelengths:

The standard spectroscopic formulations and coefficients for energy levels(4) were used to calculate the vibration-rotation state energies within the $a^3\Sigma g^+$ state and the $c^3\Pi u$ state in molecular hydrogen. Table II shows the results of these calculations. By following the $\Delta J = +1$ and $\Delta v = -1$ rules for rotation and vibration transition, and looking first at the favored J transition, i.e., J = 1 to J = 2, one readily finds a coupled set of transitions which have wavelengths matching those observed in the laboratory. These are the transitions between the energy levels shown in Table II.

TABLE II IDENTIFICATION OF THE HYDROGEN TRIPLET LASER FROM ENERGY LEVELS CALCULATED FROM PUBLISHED SPECTROSCOPIC DATA

	T_e	ω_e	$\omega_e x_e$	$\omega_e y_e$	B_e	α_e
$a^3\Sigma g^+$	95936.1	2664.83	71.65	0.920	34.216	1.671
$c^3\Pi u$	95838.5	2466.89	63.51	0.552	31.070	1.425

	TERM VALUES $a^3\Pi g^+$, J = 1	$c^3\Pi u$, J = 2	$\Delta\nu(cm^{-1})$	λ(microns)
v = 0		97238.	2601.	3.84
1	99839.	99571.	2654.	3.77
2	102225.	101782.	2699.	3.71
3	104481.			

Deuterium Energy Levels:

Assigning lines is more difficult in the case of deuterium. The problem which was straightforward for hydrogen must be worked differently for deuterium since most of the $c^3\Pi u$ state coefficients have not been determined. We must infer a set of spectroscopic coefficients which placed the observed lines in plausible transitions. In standard spectroscopic notation(4) the bottom of the potential well (electronic "zero" point) is defined by T_e, the vibration harmonics spacing by ω_e, and the anharmonic correction by $\omega_e x_e$ and $\omega_e y_e$ (and sometimes higher terms); rotational energy is characterized by coefficients B_e and α_e. The energy level term value, $T(cm^{-1})$, to this degree of approximation is:

$$T = T_e + \omega_e(v + \tfrac{1}{2}) - \omega_e x_e(v + \tfrac{1}{2})^2 + \omega_e y_e(v + \tfrac{1}{2})^3 + (B_e - \alpha_e(v + \tfrac{1}{2}))\, J(J + 1) \quad ,$$

where v and J are the vibration and rotation quantum numbers respectively. In the case of deuterium the coefficients for the $a^3\Sigma g^+$ state are known but for the $c^3\Pi u$ state values of T_e, ω_e, $\omega_e x_e$, $\omega_e y_e$, α_e and B_e are not given(4). The data we have are the three measured laser lines and the published(4) lowest lying energy level for the $c^3\Pi u$ state. With this limited data we make assumptions as follows to derive the coefficients:

a) We assumes values for Be ≈ 15. and $\alpha_e \sim .5$, values which we estimate to be within ±20% of the true value. Errors in these coefficients are least significant for J = 0, where we expect to find the transitions.

b) From the published ν_{oo}, the lowest lying level, we obtain a term value for the $c^3\Pi u$ (v = 0, J = 0) state of 96729 cm^{-1}.

c) We have assumed the $(v + \tfrac{1}{2})^3$ term, $\omega_e y_e$, is the highest correction term required so that the derivative $d^3T/d(v + \tfrac{1}{2})^3$ is a constant, 6 $\omega_e y_e$, and all higher derivatives are zero.

d) We assume a kinetics relationship identical to that of hydrogen, i.e., the three lines are in a vibrationally coupled cascade.

The problem now may be solved by solutions of simultaneous linear equations in which the values for T_e, ω_e $\omega_e x_e$ and $\omega_e y_e$ are the unknowns. Another method is to solve the problem by inspection and trial solutions. In this method the equations for T and its first three derivatives are treated as difference equations and term values are generated by assuming three adjacent vibrational transitions in the J = 0 to J = 1 series and in the J = 2 to J = 3 series. The objective is to derive values for T_e, ω_e, $\omega_e x_e$, and $\omega_e y_e$ which are plausible, i.e., Te(c) should be about 100 cm^{-1} lower than T_e(a), ω_e for "c" and "a" states should be reasonably close together, $\omega_e x_e$ should be a positive number in the range of, say, 30 to 60 cm^{-1} and $\omega_e y_e$ should preferably be approximately +1. These "requirements" only say that the $c^3\Pi u$ state of deuterium is similar to its neighboring "a" state and the whole structure is analogous to the like hydrogen states. We have tried a number of combinations the most likely two of which is shown in Table IIIA; Table IIIB shows an alternative fit which is almost as good. The best fit to the data appears to be

for the vibrational cascade starting with v = 6 and J = 0 as suggested to us by Dabrowski and Herzberg(11). In this case we determined that term coefficients for the $c^3\Pi u$ state are: T_e = 95855, ω_e = 1769, $\omega_e x_e$ = 40.88, and $\omega_e y_e$ = 1.25. These values, plus our assumed ones for B_e and α_e generate line wave numbers which are within $\pm$ 2 cm^{-1} of those derived experimentally. Our values must be considered as tentative since they are based on only four data points and show only that our data can be used to produce plausible results . Identification of additional line in this cascade, particularly of longer wavelengths would help resolve the question of where the cascade starts and other lines at different temperatures would be valuable in determining J dependence.

TABLE IIIA. IDENTIFICATION OF OUR BEST FIT TO THE DEUTERIUM TRIPLET LASER DATA

	T_e	ω_e	$\omega_e x_e$	$\omega_e y_e$	B_e	α_e
$a^3\Sigma_g^+$	95958.1	1885.84	35.96	0.340	17.109	0.606
# $c^3\Pi u$	95855.	1769.0	40.88	1.25	15.*	.5*

TERM VALUES

	$a^3\Sigma g^+$, J = 0	$c^3\Pi u$, J = 1	$\Delta\nu(cm^{-1})$	λ(microns)
v = 3		101626.	2121.	4.71
4	103747.	103127.	2172.	4.60
5	105299.	104580.	2210.	4.52
6	106790.			

* Estimate only - no temperature variation data obtained.

\# Our proposed values based on transitions suggested by I. Dabrowski and G. Herzberg (See Ref. 3).

TABLE IIIB. AN ALTERNATIVE FIT TO THE DEUTERIUM TRIPLET LASER DATA

	T_e	ω_e	$\omega_e x_e$	$\omega_e y_e$	B_e	α_e
$a^3\Sigma_g^+$	95958.1	1885.84	35.96	0.340	17.109	0.606
$c^3\Pi u$	95892.	1682.4	15.75	-0.83	15*	.5*

TERM VALUES

	$a^3\Sigma g^+$, J = 0	$c^3\Pi u$, J =1	$\Delta\nu(cm^{-1})$	λ(microns)
v = 2		100014.	2119.	4.72
3	1021.33,	101578.	2169.	4.61
4	103747.	103094.	2205.	4.54
5	105299.			

* Estimate only - no temperature variation data obtained.

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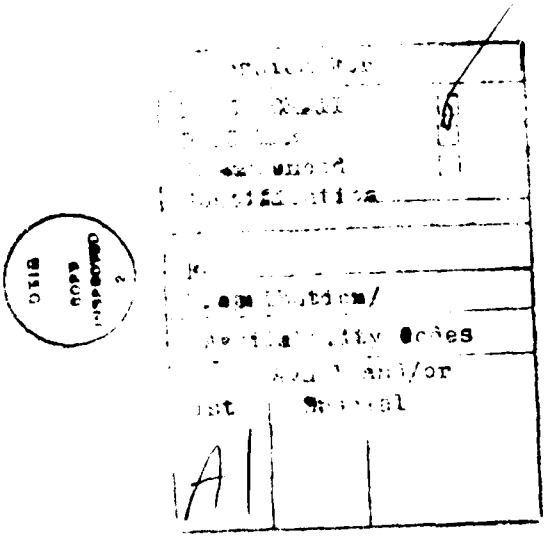

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